Experimental and theoretical characterization of \( \text{Li}_2\text{TiO}_3 \) and \( \text{Li}_4\text{SiO}_4 \) pebbles

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ABSTRACT

This paper deals with compression tests on single pebbles of \( \text{Li}_2\text{TiO}_3 \) and \( \text{Li}_4\text{SiO}_4 \) (the most promising candidates as breeder materials). Useful information have been obtained from these tests in order to assess a theoretical model developed by the authors. The experimental results have been used in order to implement a numerical model which simulates the mechanical behaviour of the pebble.

In the second part of the paper, numerical simulations of oedometer tests on pebble bed, performed with FEM codes, are illustrated. The experimental results are compared with numerical ones obtained using the Cam-Clay model. The issues derived from the use of the soil model for describing the pebble bed behaviour are described.

1. INTRODUCTION

Ceramic materials are investigated in order to develop breeding blanket modules which will be experimentally tested in the ITER reactor. Several research activities are being carried out on ceramic pebbles as well as on pebble beds with the aim to improve the technological processes, to determine the thermal and mechanical properties and to develop suitable models for simulating the behaviour.

This paper reports the results of mechanical characterization tests on Lithium Metatitanate and Lithium Orthosilicate spheres. The test is a compression uniaxial test (without radial constraints) on single spheres made of \( \text{Li}_2\text{TiO}_3 \) and \( \text{Li}_4\text{SiO}_4 \). This test allows to determine the pebble stiffness in a complete cycle of loading and unloading. The experimental results are compared with those obtained by FEM model which simulate the compression of the pebble. These simulations demonstrate the importance to know very well the material characteristic of the pebble which depend on the production technology. A numerical analysis of a pebble bed in an oedometer test is performed using a model (Cam-clay) which describes the behaviour of the soil. Choosing in appropriate manner the model parameters it is possible to fit very well the experimental results, but the physical meaning of the parameters is not straightforward.

2. EXPERIMENTAL TESTS ON SINGLE PEBBLES

Experimental compression tests were performed on Lithium-Metatitanate (\( \text{Li}_2\text{TiO}_3 \)) pebbles, 1.1-1.3 mm of diameter, and on Lithium-Orthosilicate pebbles (\( \text{Li}_4\text{SiO}_4 \)), 0.55-0.6 mm of diameter. The first pebbles were produced by CEA while the Lithium-Orthosilicate pebbles were produced by Schott-FZK. The pebbles were analyzed by SEM before the test in order to verify the actual shapes and dimensions. These examinations permitted to estimate the pebble average diameter and to normalize the results to a nominal diameter equal to 1 mm for the \( \text{Li}_2\text{TiO}_3 \) pebbles and 0.5 mm for the \( \text{Li}_4\text{SiO}_4 \) pebbles. Moreover the pebbles or their fragments (if the pebble collapsed) were again examined by SEM for evaluating the failure mode and the fracture surfaces.

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Figures 1 and 2 show the images of the pebbles obtained by the SEM and report their average diameters and the dimensions of their bounding box. The maximum variation of the average diameter of the tested pebbles is 7.8% for the Li$_2$TiO$_3$ pebbles and 4% for Li$_4$SiO$_4$ pebbles. Some pebbles were subjected to several loading cycles without reaching the failure while other pebbles were compressed until the collapse. Figures 3 and 4 illustrate for the Li$_2$TiO$_3$ pebbles the curves of the load versus displacement obtained during the loading and unloading phases, respectively. Figures 5 and 6 show the correspondent curves of the Li$_4$SiO$_4$ pebbles. The regression curves calculated with the least square method are expressed by the following equations:

a) Lithium-Metatitanate
- loading phase: \[ P = 687.55 \times s^{1.257} \] \[ r^2 = 0.9917 \]
- unloading phase: \[ P = -3.287 + 733.62 \times s \] \[ r^2 = 0.9854 \]

b) Lithium-Orthosilicate
- loading phase: \[ P = 3613 \times s^{1.9027} \] \[ r^2 = 0.927 \]
- unloading phase: \[ P = 7657 \times s^{1.8922} \] \[ r^2 = 0.974 \]
where $P$ (N) is the applied load, $s$ (mm) is the vertical displacement and $r^2$ is the correlation coefficient.

Before the regression analyses, the values of the experimental displacement were normalized at the nominal diameter assuming according the Hertz theory that the displacement are inversely proportional to the pebble radius.
Even though the number of the pebble tested is not statistically significative, nevertheless the number of the collapsed pebbles versus the maximum load could give an useful information on the ultimate strength of the pebbles. Figures 7 and 8 show the fraction of the collapsed pebbles versus the maximum load for the Li$_2$TiO$_3$ and Li$_4$SiO$_4$ pebbles, respectively. No Li$_2$TiO$_3$ pebbles collapsed under 25 N while 1 N is the maximum load supported by the Li$_4$SiO$_4$ pebbles without any rupture (Indeed just only one pebble collapsed at 1.06 N while all the other broke for load upper than 7 N.

3. NUMERICAL SIMULATION OF THE PEBBLE COMPRESSION

The compression tests on single pebbles have been simulated numerically by means of the FEM code MSC-MARC [4]. The aims of the simulations were to implement a model which described correctly the behaviour of the pebble comparing the results with the experimental ones as well as to understand the main phenomena which occur during the pebble compression.
Figure 9 shows the implemented mesh. The mesh is made up of axial symmetric elements. The load is applied by means of a rigid upper plate and the pebble is located on a rigid lower plate. The constraints between the pebble and the plates have been simulated by means of unilateral contacts (reacting only to compression loads) without friction.

Being the pebble made of a ceramic material having a low tension resistance but a great compression strength, the implemented model assumes a linear elastic behaviour under tensile stress and an elastic perfectly plastic behaviour under compressive stress. The material fails if the tensile stress overcomes the cracking stress value ($\sigma_{cr}$) or if the plastic strain (under compressive stresses) reaches the crushing strain value ($\varepsilon_{u}$). Other parameters which characterize the material behaviour are:

- the softening module ($E_s$), which represents the slope of the curve $\sigma$-$\varepsilon$ after the cracking stress (that is, the manner in which the stress goes to zero),
- the shear retention ($\tau$), which means the capability of the material to transmits shear by friction if the crack closure occurs.

The material data for the lithium metatitanate and Lithium Ortosilicate are reported in [1]- [3] and are summarize as follows:

- Lithium-Metatitanate : $E= 200.6$ GPa - $\nu = 0.27$ - $\sigma_{u}= 1113$ MPa
- Lithium-ortosilicate : $E= 90$ GPa - $\nu = 0.25$ - $\sigma_{u}= 880$ MPa

where $E$ is the Young module, $\nu$, the Poisson coefficient and $\sigma_{u}$ the ultimate compression stress.

Using these data for obtaining the material parameters, the numerical results do not fit the experimental ones. $E$ and $\sigma_{u}$ seem too high values. In fact considering the average stresses and strains obtained in the single pebble compression tests, the Young modules have to be two or three order of magnitude lower than the above mentioned values. The data reported in [1]- [3] are data collected from different sources and they could not match the material characteristics of the tested pebbles. Indeed for the ceramic material, the thermal mechanical characteristics are strongly dependent on the production technology.

This problem was overcome performing a parametric analyses varying the model parameters in manner that the load- displacement curves fitted the experimental ones.

Figures 10 and 11 compare the experimental load-displacement curves of the lithium metatitanate and Lithium Ortosilicate with those obtained numerically. In Fig.10, an experimental cycle on a Lithium-
Metatitanate pebble (sample T22) fits the numerical results with the following values of the material parameters: \( E = 5 \text{ GPa}; \sigma_y = 200 \text{ MPa}; \varepsilon_u = 0.15; \sigma_{cr} = 100 \text{ MPa}; \) \( E_s = 0.2 \text{ MPa}. \) In the same Figure a cycle built with the regression curves fits the numerical curve assuming \( E = 5 \text{ GPa} \) and \( \varepsilon_u = 0.3. \)

Figure 11 shows the comparison of three cycles of loading and unloading on a Lithium-ortosilicate pebble (sample T44) and the correspondent curves obtained by means of the numerical simulation. A good agreement is obtained assuming the following values of the material parameters:

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E = 5.5 \text{ GPa}; \sigma_y = 250 \text{ MPa}; \varepsilon_u = 0.15; \sigma_{cr} = 139 \text{ MPa}; \] \( E_s = 0.2 \text{ MPa}. \)

Figure 12 illustrates the diagrams of the vertical stress (with the sign changed) versus the total equivalent strain for three points of the pebble, located at different distance from the contact zone (0.0125 mm, 0.1 mm and 0.5 mm). The simulation corresponds to the loading phase of the cycle in Fig.10 compared to the regression curve. On the pebble centre the vertical stress and the strain are 4 times lower than near the contact zone.

For the same case, Figure 13 shows the distribution of the plastic strains and Von Mises stresses on the Lithium Metatitanate pebble. A great part of the pebble is yielded for a load of 47 N. The maximum plastic strain is 0.19.

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**Fig. 13 – Distribution of the plastic strain and Von Mises stress on the Lithium Metatitanate pebble**

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4 NUMERICAL SIMULATION OF A LITHIUM ORTOSILICATE PEBBLE BED WITH AN HOMOGENEOUS MODEL

Several Authors simulate the pebble bed behaviour by means FEM codes using the model developed for the soil (Cap model of Drucker and Prager[5], Cam-clay model of Roscoe and Burland [6] and so on). These model depend on several soil parameters which have not a precise correspondence with the mechanical...
characteristics of the pebbles. In the following a numerical simulation of an oedometer test performed on Lithium-ortosilicate pebble bed is illustrated. This simulation is performed using the MSC-Marc FEM code and assuming that the pebble bed behaviour is described by means of the Cam-clay model. The numerical results (vertical load versus vertical displacement) are compared with the experimental ones obtained by the Authors of this paper [7]. The Cam-clay model depends on several parameters. In order to find a best fit between the numerical and experimental results we have varied the main parameters in reasonable ranges.

The main parameters of the Cam-clay model are the following:
- \( M \), the slope of the critical state line
- \( p_c \), preconsolidation pressure (a stress-like hardening variable)
- \( K \), the slope of the over consolidation lines
- \( \lambda \), the slope of the normal consolidation line.

\( M \) and \( p_c \) enter in the yield criteria while \( K \) and \( \lambda \) enter in the equation which describe the isotropic compression of the soil (\( K \) in the elastic range and \( \lambda \) in the elastic plastic range). The isotropic compression

![Fig.14 – Influence of the parameter \( \lambda \)](image1)

![Fig.15 – Influence of the parameter \( p_c \)](image2)

![Fig.16 – Influence of the parameter \( k \)](image3)

![Fig.17 – Influence of the parameter \( M \)](image4)
constitutive relations give the specific volume (ratio between the total volume of the soil and the volume of the solid part) as function of the logarithm of the pressure.

No influence have in the Cam-clay model the Young module, the Poisson coefficient and the yielding stress. A good agreement between the experimental curve and numerical one was obtained assuming the following values for the four parameters: \( M=0.5; \ K=0.002; \ \lambda=0.006; \ \rho_c=0.9 \) MPa.

Figures 14-17 shows the influence of the different parameters on the solution assumed as the best. Obviously several other groups of variable could give a solution which agrees with the experimental curve. A big question is the possibility of the extrapolation of the simple model of the oedometer test at more complex stress states. The stresses in different part of the pebble are very different (Fig.13) but the homogeneous model of the oedometer test gives everywhere a constant stress and a constant strain. It is not easy to relate this effective stress to the actual stress of the pebble.

5. CONCLUSIONS

This paper illustrated the experimental results of compression tests on single pebbles. The tests permitted to estimate the fracture load of Lithium-Metatitanate pebbles (1.1–1.3 mm of diameter) and of Lithium-Orthosilicate pebbles (0.55–0.6 mm of diameter). The numerical simulations of the pebble compression give results which agree very well with the experimental ones assuming values of the material characteristics different from those found in the technical literature for these materials. This fact could be explained considering that the mechanical characteristics of ceramic materials depend strongly on the production technology.

An homogeneous model developed for soil has been applied to simulate an oedometer test of a Lithium-Orthosilicate pebble bed. Choosing appropriate values for the several constants it is possible to fit the experimental curve, but the physical meaning of the parameter values is not immediately extrapolate to the pebble bed.

REFERENCES